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Article

Density, Speed of Sound, Viscosity, and Surface Tension of Hexamethylenetetramine + Water + Ethanol Ternary Mixtures from T = 293.15 to 323.15 K

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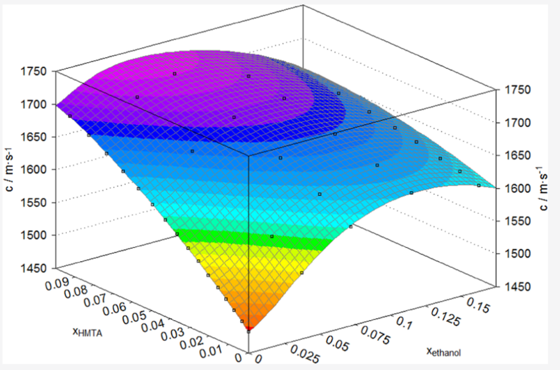
ABSTRACT: This research work analyzes the behavior of ternary mixtures composed of hexamethylenetetramine, water, and ethanol in relation to the influence of composition and temperature on several physical properties such as density, speed of sound, viscosity, and surface tension. The presence of ethanol in aqueous solutions of hexamethylenetetramine causes important changes in the behavior of the liquid mixture in comparison to blends in the absence of ethanol. One of the most important effects is the elimination of the presence of temperature-resistant structures (clathrate type) only with low additions of ethanol.



INTRODUCTION

Hexamethylenetetramine (HMTA) is a chemical compound with very important uses at the industrial level (fine chemical and industrial processes) such as personal care formulations, as a curing agent in rubber and epoxy production, and as an additive in adhesives and food preservative.[1] It is also widely employed as a catalyst and a multifunctional ligand in important chemical reactions with interest in industry.[2−7] In relation to the last type of applications, the interactions produced by HMTA takes an special interest by the existence of hydrogen bond-type strong interactions. Also, this chemical compound has shown suitable solubility in aqueous solutions and using polar organic solvents.[6]

Previous studies related with the characterization of aqueous solutions of HMTA are reduced only to initial works centered on the determination of the effect of composition on density and viscosity at 298.15 K[7] and the influence of temperature in the range of 276.15−307.15 K.[8] These previous studies were extended by our research team analyzing other physical properties using wider composition and temperature ranges for HMTA + water mixtures.[9] This last study allows us to confirm the presence of temperature-resistant structures that were detected by the characteristic behavior of temperature and composition on speed of sound experimental data.

This work analyzes the behavior of HMTA mixtures using blends of water and ethanol as solvent. Ethanol has been chosen taking into account the use of this compound as a solvent or co-solvent in chemical reaction, where HMTA is employed as a catalyst[10] or ligand.[11] On the basis of the possible strong interaction of HMTA with the solvent, the present work is focused on the evaluation of several physical properties and the influence of the mixture composition to evaluate its influence on transport properties and the relation with the chemical interactions.

EXPERIMENTAL SECTION

Materials. Information of the reagents used in the present work is presented in Table 1. HMTA and ethanol have been employed without further purification. Distilled water has been employed to prepare different ternary mixtures. Liquid mixtures were prepared by mass using an analytical balance (Mettler Toledo ME-T).

METHODS

Density and Speed of Sound. The experimental values of density (ρ) and speed of sound (c) of samples were determined using an Anton Paar DSA 5000 vibrating tube densimeter and a sound analyzer. Air and water were used for calibration. The transducer emits sound waves at a frequency

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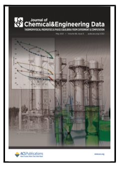
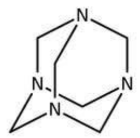
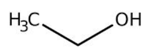


Table 1. Sample Description Table

chemical name	CAS number	molar mass (g·mol-1)	source	mole fraction purity	analysis method
HMTA[a] 	100-97-0	140.19	Sigma-Aldrich	≥ 0.99	GC[b]
Ethanol 	64-17-5	46.07	Sigma-Aldrich	≥ 0.99	Titration[c]

[a]Hexamethylenetetramine. [b]Gas chromatography, from vendor certificate of analysis. [c]Titration with NaOH, from vendor certificate of analysis.

of 3 MHz. The temperature range employed in the present work was 293.15−323.15 K.

Viscosity. The experimental values corresponding to the kinematic viscosity (ν) of liquid samples were calculated using the transit time of the liquid meniscus through Ubbelohde viscosimeters with several capillaries (Schott). Capillaries Ia (K = 0.05047 $mm^2 \cdot s^{-2}$) and I (K = 0.01013 $mm^2 \cdot s^{-2}$) have been employed in a Schott-Geräte AVS 350 viscosimeter. Equation 1 was employed to calculate the viscosity

$$\nu = K \cdot (t - \theta) \quad (1)$$

where t is the efflux time; K is the characteristic constant of the capillary viscosimeter; and θ is a coefficient to correct end effects. Correction coefficient was calculated using data from the supplier (Schott). An electronic stopwatch was used to measure efflux times. The average value of five consecutive measurements was employed. The dynamic viscosity (η) was calculated from the product of the kinematic viscosity (ν) and density (ρ) using eq 2.

$$\eta = \nu \cdot \rho \quad (2)$$

Surface Tension. Surface tension (σ) experimental data were obtained using a Krüss K-11 tensiometer by the Wilhelmy plate method. The plate employed was a platinum plate supplied by Krüss. This plate was cleaned and flame-dried before each experimental measurement. Five measurements were employed to determine each surface tension value. Samples were thermostatted in a closed stirring vessel before each measurement.

■ RESULTS AND DISCUSSION

Tables 2−5 show the experimental data of density, viscosity, speed of sound, and surface tension of ternary mixtures of water + ethanol + HMTA in the solubility range, previously determined in the literature,[12,13] and using different temperatures (between 293.15 and 323.15 K).

In a previous work,[9] the influence of HMTA in aqueous solution on different physical properties was studied concluding that an increase in the presence of this amine causes a monotonic increase in density. This behavior is included in Figure 1. On the other hand, this figure also includes the effect of the presence of ethanol on density when the water−ethanol mixture is analyzed. The behavior showed for this last blend is the opposite of the previous one commented (HTMA + water). When part of water is replaced by different amounts of ethanol using a ternary mixture (water + ethanol + HMTA) in the solvent, the trend observed for density is similar to the behavior corresponding to the water + HMTA mixture. It involves an increase in density with HMTA concentration. A higher presence of ethanol in the ternary mixture causes a decrease in the value of density.

A change in temperature does not modify the shape of the curve corresponding to the influence of HMTA concentration on density using different water/ethanol ratios. An increase in temperature causes a decrease in density that is a behavior commonly observed in aqueous solutions of amines. The direct influence of temperature on density shows a decrease with a linear trend and a relatively constant value for the slope.

Aqueous solutions of HMTA + water showed a characteristic behavior with the formation of certain type of structures (clathrates). This behavior consists of a lack of influence of temperature on some physical properties. One of them is the speed of sound that allows us to characterize this type of mixtures. The behavior corresponding to the HMTA + water binary mixture is shown in Figure 2 using solid and dashed lines for speed of sound experimental data at 293.15 and 323.15 K, respectively.[9] At a low HMTA mole fraction, speed of sound data are lower for lower temperatures. An increase in HMTA concentration causes a decrease in the effect of temperature on experimental data until the same value is reached at an HMTA molar fraction close to 0.08. This behavior has been observed only for some amines in aqueous solutions[16−18] related with the formation of rigid structures and especially clathrates for this type of aqueous solution.[19]

Figure 2 also includes the same type of plot using experimental data corresponding to the influence of HMTA composition on speed of sound for ternary mixtures and using different water/ethanol ratios. When low amounts of ethanol are used in the mixture, a similar behavior is obtained in comparison to HMTA + water blends. For the ternary mixtures with low ethanol additions, the crossing point is reached at lower amine concentrations than the mixture in the absence of ethanol.

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Table 2. Density ρ, Speed of Sound c, Dynamic Viscosity η, and Surface Tension σ of HMTA (1) + Water (2) + Ethanol (3) from T = 293.15 to 323.15 K Using a Water/Ethanol Mass Ratio of 2 at $p = 10^5$ Pa[a]

x_1	x_3	T (K) 293.15	T (K) 303.15	T (K) 313.15	T (K) 323.15
		ρ (g·cm^{-3})			
0.000	0.163	0.9469	0.9404	0.9354	0.9310
0.009	0.162	0.9653	0.9587	0.9517	0.9443
0.018	0.160	0.9774	0.9708	0.9637	0.9563
0.029	0.158	0.9924	0.9857	0.9786	0.9712
0.039	0.157	1.0044	0.9976	0.9905	0.9830
0.051	0.155	1.0165	1.0097	1.0025	0.9951
0.065	0.153	1.0312	1.0243	1.0172	1.0097
		c (m·s^{-1})			
0.000	0.163	1611.4	1587.5	1563.9	1539.1
0.009	0.162	1621.4	1598.3	1574.6	1550.4
0.018	0.160	1630.8	1607.7	1584.0	1559.7
0.029	0.158	1642.9	1619.5	1595.4	1570.6
0.039	0.157	1652.2	1628.8	1604.8	1579.7
0.051	0.155	1661.9	1638.7	1614.6	1589.5
0.065	0.153	1673.3	1649.7	1625.2	1599.9
		η (mPa·s)			
0.000	0.163	2.655	1.873	1.350	1.004
0.009	0.162	3.015	2.102	1.515	1.131
0.018	0.160	3.447	2.388	1.711	1.272
0.029	0.158	4.047	2.807	2.082	1.556
0.039	0.157	4.669	3.237	2.319	1.749
0.051	0.155	5.372	3.630	2.571	1.892
0.065	0.153	6.477	4.399	3.108	2.289
		σ (mN·m^{-1})			
0.000	0.163	33.5	32.0	30.6	29.3
0.009	0.162	33.6	32.1	30.7	29.4
0.018	0.160	33.7	32.4	31.0	29.6
0.029	0.158	34.0	32.8	31.4	30.1
0.039	0.157	34.3	33.1	31.8	30.5
0.051	0.155	34.8	33.7	32.4	31.2
0.065	0.153	35.9	34.7	33.5	32.2

[a]Standard uncertainties u are $u(T)$ = 0.01 K, $u(p)$ = 2 kPa, and $u(x)$ = 0.001, and the combined expanded uncertainties U_c (level of confidence = 0.95, k = 2) are $U_c(\rho) = 3 \times 10^{-3}$ g·cm^{-3}, $U_c(c)$ = 0.5 m·s^{-1}, $U_c(\eta)$ = 0.04 mPa·s, and $U_c(\sigma)$ = 0.3 mN·m^{-1}.

When lower water/ethanol ratios are used, this higher presence of ethanol avoids the formation of this type of temperature-resistant structures observing higher speed of sound experimental data for the lower temperature over the entire HTMA concentration range.

The analysis of the influence of composition in binary and ternary mixtures on viscosity allows us to obtain the behaviors included in Figure 3. Ternary mixtures have shown a similar behavior in relation to the shape of the curve of the influence of amine concentration on viscosity. In general, a monotonic increase in viscosity was observed with an increase in amine concentration. The presence of different amounts of ethanol does not modify the shape of the curve but causing an increase in the magnitude of this property. The effect of amine seems important in the ternary mixture and similar to the corresponding one for the HMTA + water binary blend.

The increase in viscosity with ethanol concentration is in agreement with the previously observed behavior for the binary mixture of ethanol + water[14] due to the high interaction between both molecules.

The influence of temperature on the viscosity for ternary and binary mixtures involving HMTA can be observed in Figure 4. The presence of temperature-resistant structures (observed for systems without ethanol and ternary mixtures with low ethanol composition) does not show a clear effect on viscosity in comparison to the other mixtures without the presence of this type of rigid structures. This lack of influence is in agreement with previous studies with similar type of mixtures.[22]

The influence of temperature on the value of viscosity for binary and ternary mixtures has shown a monotonic decrease (see Figure 4) that is commonly observed for this type of aqueous mixtures with amines with a Newtonian behavior. This influence has been fitted using the fluidity concept as in eq 3.[23]

$$\left(\frac{1}{\eta}\right)^{\phi} = a + b \cdot T \tag{3}$$

where a and b are liquid-phase characteristic parameters and ϕ is the fluidity exponent that depends on the nature of the liquid. Several studies have obtained average values for fluidity exponent (0.312 for alcohols and 0.300 for ionic liquids).[23,24] For the mixtures employed in the present work, the fluidity equation shows a suitable behavior, and an average value for fluidity exponent of 0.244 ± 0.018 was obtained.

Figure 5 shows the observed behavior of this type of mixtures in relation to surface tension experimental data. It shows a comparison between experimental values obtained for the ternary mixtures (HMTA + ethanol + water) and the corresponding ones for aqueous solutions of HMTA and ethanol.

The behaviors corresponding to binary mixtures (HMTA + water and ethanol + water) and included in Figure 5 show a completely different trend from the corresponding one for the ternary mixtures. Aqueous solutions of HMTA show a decrease in the value of surface tension with the increase in amine concentration until a relatively constant value is reached. This type of behavior has been commonly observed for aqueous solutions of amines[17,25] though the magnitude of the decrease in the surface tension is in general higher than that observed for the HMTA + water blend.

The other binary mixture (ethanol + water) has also shown a decrease in the value of this physical property with the increase of ethanol composition. In the studied composition range, a constant value was not reached for this mixture, but the decrease in comparison to the use of HMTA was higher.

In relation to the influence of composition for the ternary mixtures on surface tension, the experimental data show that low composition of ethanol and HMTA maintains a similar behavior to the previously described ones for binary mixtures that is a decrease in surface tension. An increase in amine concentration causes a change in this type of behavior, producing also an increase in surface tension. This effect caused by amine is observed for all of the ternary mixtures with a certain value of ethanol concentration. In fact, when ethanol concentration is increased, the initial decrease in the value of surface tension at a low amine concentration is not observed and only a monotonic increase in this property is observed.

The influence of temperature on surface tension for these mixtures shows a linear trend (see Figure 4) for all of the ternary mixtures employed in the present work. The slope of this type of plot has shown a relatively constant value independent of mixture composition. For ternary mixtures

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Table 3. Density ρ, Speed of Sound c, Dynamic Viscosity η, and Surface Tension σ of HMTA (1) + Water (2) + Ethanol (3) from T = 293.15 to 323.15 K Using a Water/Ethanol Mass Ratio of 3 at $p = 10^5$ Pa[a]

x_1	x_3	T (K) 293.15	T (K) 303.15	T (K) 313.15	T (K) 323.15
		ρ (g·cm^{-3})			
0.0000	0.1152	0.9597	0.9547	0.9508	0.9463
0.0164	0.1133	0.9892	0.9836	0.9774	0.9708
0.0365	0.1110	1.0154	1.0095	1.0031	0.9963
0.0609	0.1082	1.0414	1.0351	1.0285	1.0214
0.0779	0.1063	1.0582	1.0517	1.0450	1.0379
		c (m·s^{-1})			
0.0000	0.1152	1627.3	1611.4	1594.0	1575.3
0.0164	0.1133	1649.5	1632.5	1614.2	1595.3
0.0365	0.1110	1673.1	1655.1	1635.2	1614.7
0.0609	0.1082	1696.9	1677.6	1657.0	1634.9
0.0779	0.1063	1710.0	1689.9	1667.9	1645.2
		η (mPa·s)			
0.0000	0.1152	2.348	1.647	1.198	0.902
0.0164	0.1133	3.014	2.117	1.555	1.169
0.0365	0.1110	4.124	2.785	2.112	1.600
0.0609	0.1082	5.918	4.023	2.898	2.157
0.0779	0.1063	7.432	5.034	3.533	2.596
		σ (mN·m^{-1})			
0.0000	0.1152	36.0	35.0	33.8	32.6
0.0164	0.1133	36.6	35.6	34.5	33.5
0.0365	0.1110	37.8	36.9	35.9	35.0
0.0609	0.1082	39.1	38.2	37.2	36.3
0.0779	0.1063	39.8	38.9	37.9	36.9

[a]Standard uncertainties u are $u(T)$ = 0.01 K, $u(p)$ = 2 kPa, and $u(x)$ = 0.001, and the combined expanded uncertainties U_c (level of confidence = 0.95, k = 2) are $U_c(\rho) = 3 \times 10^{-3}$ g·cm^{-3}, $U_c(c)$ = 0.5 m·s^{-1}, $U_c(\eta)$ = 0.04 mPa·s, and $U_c(\sigma)$ = 0.3 mN·m^{-1}.

Table 4. Density ρ, Speed of Sound c, Dynamic Viscosity η, and Surface Tension σ of HMTA (1) + Water (2) + Ethanol (3) from T = 293.15 to 323.15 K Using a Water/Ethanol Mass Ratio of 5 at $p = 10^5$ Pa[a]

x_1	x_3	T (K) 293.15	T (K) 303.15	T (K) 313.15	T (K) 323.15
		ρ (g·cm^{-3})			
0.0000	0.0725	0.9724	0.9692	0.9657	0.9602
0.0157	0.0713	0.9992	0.9948	0.9897	0.9839
0.0348	0.0700	1.0253	1.0204	1.0149	1.0090
0.0578	0.0683	1.0516	1.0462	1.0403	1.0339
0.0868	0.0662	1.0788	1.0729	1.0666	1.0600
		c (m·s^{-1})			
0.0000	0.0725	1600.8	1598.8	1593.9	1585.9
0.0157	0.0713	1630.4	1625.6	1618.1	1607.4
0.0348	0.0700	1662.3	1654.1	1643.3	1630.1
0.0578	0.0683	1695.4	1683.5	1669.8	1654.6
0.0868	0.0662	1725.1	1709.5	1692.5	1673.7
		η (mPa·s)			
0.0000	0.0725	1.854	1.336	0.995	0.747
0.0157	0.0713	2.453	1.765	1.314	1.007
0.0348	0.0700	3.462	2.468	1.786	1.372
0.0578	0.0683	5.095	3.524	2.548	1.921
0.0868	0.0662	7.814	5.313	3.769	2.781
		σ (mN·m^{-1})			
0.0000	0.0725	40.3	39.2	38.3	37.3
0.0157	0.0713	39.3	38.4	37.2	36.2
0.0348	0.0700	41.1	40.2	39.2	38.2
0.0578	0.0683	43.4	42.5	41.3	40.4
0.0868	0.0662	46.0	45.1	44.2	43.2

[a]Standard uncertainties u are $u(T)$ = 0.01 K, $u(p)$ = 2 kPa, and $u(x)$ = 0.001, and the combined expanded uncertainties U_c (level of confidence = 0.95, k = 2) are $U_c(\rho) = 3 \times 10^{-3}$ g·cm^{-3}, $U_c(c)$ = 0.5 m·s^{-1}, $U_c(\eta)$ = 0.04 mPa·s, and $U_c(\sigma)$ = 0.3 mN·m^{-1}.

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Table 5. Density ρ, Speed of Sound c, Dynamic Viscosity η, and Surface Tension σ of HMTA (1) + Water (2) + Ethanol (3) from T = 293.15 to 323.15 K Using a Water/Ethanol Mass Ratio of 10 at p = 10^5 Pa[a]

x_1	x_3	T (K) 293.15	T (K) 303.15	T (K) 313.15	T (K) 323.15
		ρ (g·cm^{-3})			
0.0000	0.0376	0.9842	0.9811	0.9772	0.9726
0.0151	0.0370	1.0084	1.0049	1.0006	0.9957
0.0329	0.0364	1.0325	1.0282	1.0230	1.0189
0.0552	0.0355	1.0577	1.0552	1.0512	1.0467
0.0830	0.0345	1.0832	1.0800	1.0753	1.0700
		c (m·s^{-1})			
0.0000	0.0376	1550.5	1562.9	1570.1	1573.0
0.0151	0.0370	1587.0	1594.9	1598.4	1597.8
0.0329	0.0364	1626.7	1625.2	1620.0	1618.0
0.0552	0.0355	1669.4	1662.7	1654.9	1648.8
0.0830	0.0345	1713.3	1704.7	1694.0	1680.5
		η (mPa·s)			
0.0000	0.0376	1.412	1.048	0.783	0.585
0.0151	0.0370	1.795	1.280	0.998	0.732
0.0329	0.0364	2.550	1.745	1.325	1.000
0.0552	0.0355	3.996	2.754	2.042	1.539
0.0830	0.0345	6.675	4.613	3.334	2.511
		σ (mN·m^{-1})			
0.0000	0.0376	49.7	47.6	45.5	43.4
0.0151	0.0370	40.8	38.6	36.7	34.5
0.0329	0.0364	41.9	39.7	37.7	35.7
0.0552	0.0355	45.0	42.7	41.1	38.5
0.0830	0.0345	47.5	45.4	43.4	41.4

[a]Standard uncertainties u are $u(T)$ = 0.01 K, $u(p)$ = 2 kPa, and $u(x)$ = 0.001, and the combined expanded uncertainties U_c (level of confidence = 0.95, k = 2) are $U_c(\rho)$ = 3 × 10^{-3} g·cm^{-3}, $U_c(c)$ = 0.5 m·s^{-1}, $U_c(\eta)$ = 0.04 mPa·s, and $U_c(\sigma)$ = 0.3 mN·m^{-1}.

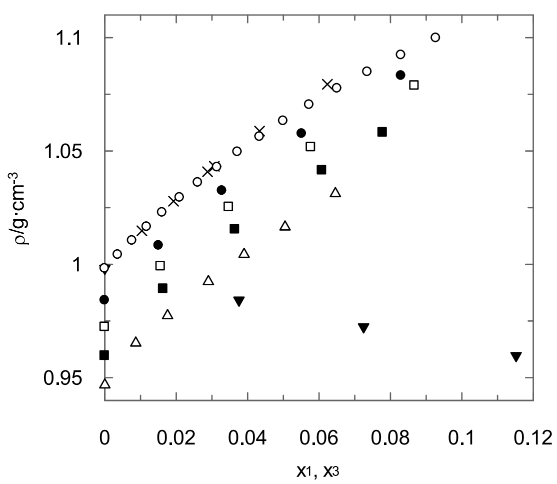

Figure 1. Effect of HMTA (1) and ethanol (3) composition on density. HTMA (1) + water (2) + ethanol (3) system: ●, W/E = 10; □, W/E = 5; ■, W/E = 3; △, W/E = 2. HMTA (1) + water (2) system: ○;[9] ×.[14] Water (2) + ethanol (3) system: ▼.[15] T (K) = 293.15.

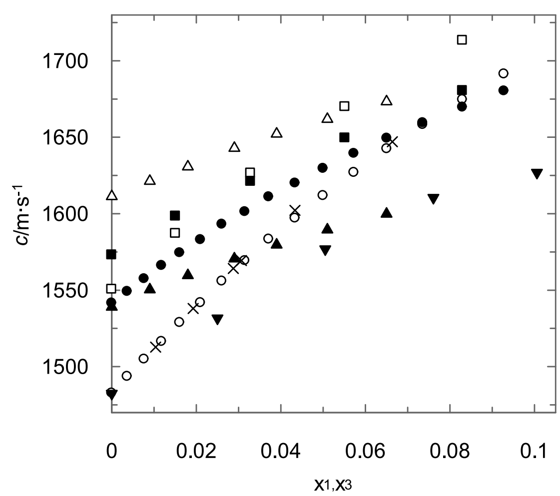

Figure 2. Influence of temperature and composition on speed of sound experimental data. HTMA (1) + water (2) + ethanol (3) system: □, W/E = 10 at T (K) = 293.15; ■, W/E = 10 at T (K) = 323.15; △, W/E = 2 at T (K) = 293.15; ▲, W/E = 2 at T (K) = 323.15. HMTA (1) + water (2) system: ○, T (K) = 293.15;[9] ●, T (K) = 323.15;[9] ×, T (K) = 293.15.[14] Water (2) + ethanol (3) system: ▼.[20] T (K) = 293.15.

analyzed in the present work, an average value of −0.098 mN·m^{-1}·K^{-1} was found for the slope. A lower slope value was observed in comparison to the mixture in the absence of ethanol[9] (−0.143 mN·m^{-1}·K^{-1}) that showed a similar value to other binary systems such as tetramethylenediamine + water[27] (−0.148 mN·m^{-1}·K^{-1}).

■ CONCLUSIONS

This research work analyzes the influence of the presence of hexamethylenetetramine in solvents based in mixtures of ethanol and water on several physical properties with high importance in the evaluation of molecular interactions and the behavior of different industrial processes.

In comparison to systems in the absence of ethanol, the presence of this co-solvent does not produce significant changes in density and viscosity. The same type of trends has

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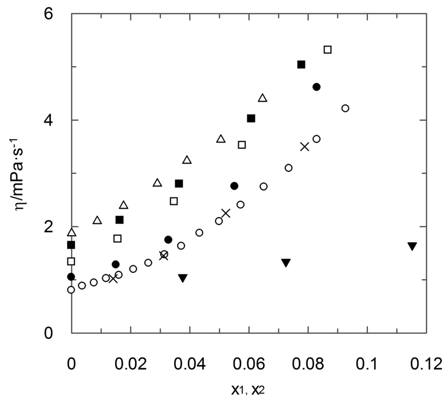

Figure 3. Influence of ternary mixture composition on dynamic viscosity. HTMA (1) + water (2) + ethanol (3) system: ●, W/E = 10; □, W/E = 5; ■, W/E = 3; △, W/E = 2. HMTA (1) + water (2) system: ○;[9] ×.[21] Water (2) + ethanol (3) system: ▼.[15] T (K) = 303.15.

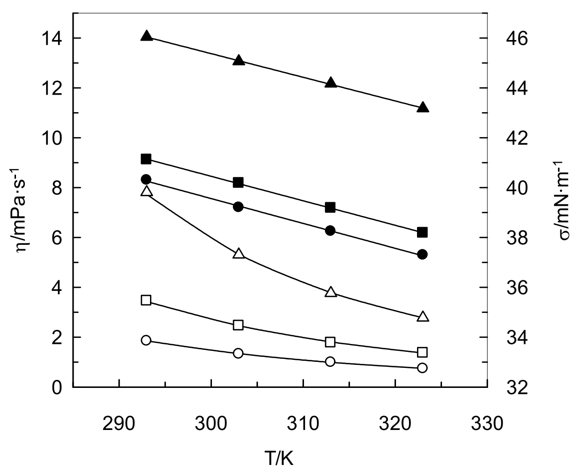

Figure 4. Effect of temperature on dynamic viscosity and surface tension. Open symbols: viscosity; full symbols: surface tension; ○ and ●, x_1= 0; □ and ■, x_1= 0.0348; △ and ▲, x_1= 0.0868. W/E = 5. Solid lines for viscosity correspond to fluidity equation.

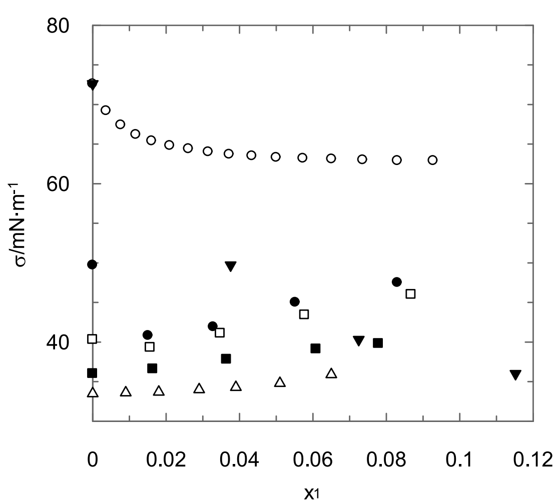

Figure 5. Influence of mixture composition on surface tension data. HTMA (1) + water (2) + ethanol (3) system: ●, W/E = 10; □, W/E = 5; ■, W/E = 3; △, W/E = 2. HMTA (1) + water (2) system: ○.[9] Water (2) + ethanol (3): ▼.[26] T (K) = 293.15.

been observed: a decrease in the value of density and an increase for viscosity.

In relation to the other properties such as speed of sound and surface tension, the effect caused by ethanol is important and causes significant changes in the overall behavior. For speed of sound data, the presence of ethanol in the mixture causes an important effect avoiding the formation of clathrates (temperature-resistant structures) with low addition of this co-solvent.

For the other property (surface tension), the presence of ethanol generates a change in the effect caused by the amine in aqueous solution. An increase in HMTA causes a decrease in surface tension in mixtures without ethanol, and the opposite behavior is observed when this co-solvent is included in the liquid mixture.

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Notes

The authors declare no competing financial interest.

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